

Pilot-scale experiments on a hybrid membrane-electrocoagulation system to produced water treatment in a domestic oil reservoir

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Abstract

The goal of this paper is to design and set up a new hybrid treatment system for purification of Iranian offshore oil company wastewater. The treatment system consists of electrocoagulation, microfiltration and reverse osmosis (RO) sections. Treatment performance is evaluated by measuring electrical conductivity (EC), chemical oxygen demand (COD), biochemical oxygen demand (BOD₅), total suspended solids (TSS), total dispersed solids (TDS) oil and grease (O&G), total hardness (TH) and turbidity. Results showed that the maximum removal efficiency of BOD₅, COD, TSS, O&G, EC, TH and turbidity were obtained as 94.49%, 92.17%, 88.18%, 80.8%, 89.84%, 86.66% and 71.72%, respectively. DO was increased significantly due to the reduction of BOD, COD and microorganisms. Results showed that the combination of iron and aluminium electrodes can be very effective in removing pollutants. In addition, the use of microfiltration before RO led to a significant reduction in salinity and TDS. Response Surface Methodology (RSM) was used to determine the optimal experimental conditions for COD, BOD₅ and turbidity using Design Expert11. RSM results showed that the optimal electric current and time for BOD₅ and COD removal is 20A and 300 min, respectively. The optimal electric current and time for turbidity were obtained as 20A and 210 min.

Key words: electrocoagulation, filtration, hybrid treatment system, produced water, wastewater treatment

Highlights

- In the electrocoagulation section, iron and aluminum were applied as electrodes with the electric current of 15A, 20A and 25A.
- The maximum DO was achieved as 6.6 mg/lit.
- The designed hybrid system has a great performance and treated water has acceptable quality.

INTRODUCTION

Oil and gas are the most important sources of energy in the world. [Figure 1](#) shows a schematic of a typical oil reservoir. In oil and gas reservoirs, water is found together with hydrocarbons. The pressure of the reservoir is reduced by extraction of oil and gas, which leads to a decrease in oil recovery. In order to maintain the hydraulic pressure of the reservoir as well as enhancing oil recovery, a significant amount of water is injected into the reservoir. Therefore, a large amount of water is extracted during extracting oil and gas, which is known as produced water. Investigations have shown that generally the oil/water ratio is 1:3 in produced water ([Fakhru'l-Razi et al. 2009](#)). Produced water is a complex of a wide range of organic and inorganic material such as dissolved and dispersed oils, grease, salts, formation solids, treating chemicals, radionuclides, waxes, heavy metals, dissolved gases, scale products, microorganisms and dissolved oxygen ([Hayes & Arthur 2004](#); [Sirivedhin et al. 2004](#); [Ray & Engelhardt 2012](#)). A large

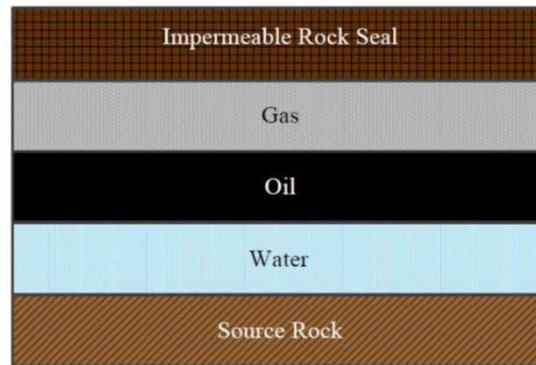


Figure 1 | Schematic of a typical reservoir.

amount of produced water is discharged into the environment, which can lead to a lot of acute environmental issues. Hence it is necessary to purify produced water before discharging into the environment. In many cases, it is also economical to reuse treated produced water in various process of the oil industry. Several purification methods are available for produced water treatment. The choice of treatment method depends on the concentration of the chemical compound in the produced water. The concentrations of water pollutants are varied in different produced waters. Generally, using traditional methods of produced water purification such as chemical, physical and mechanical methods, it is not possible to remove large quantities of aliphatic and aromatic hydrocarbons (Saien & Nejati 2007). On the other hand, the use of new methods such as membrane processes, ultrafiltration, microfiltration, nanofiltration and electrocoagulation, can lead to a significant enhancement in the quality of produced water treatment (Meyers & Meyers 2004). Ultrafiltration (UF) is an important membrane method in produced water treatment, which is usually used when soluble molecules are about ten times larger than solvent molecules (Vieira *et al.* 2001; Barakat & Schmidt 2010; Katsou *et al.* 2011). Microfiltration (MF) is generally applied as a pre-treatment for separation processes such as UF and reverse osmosis (RO). The typical particle size used for MF ranges is from about 0.1–10 μm (Song *et al.* 2006; Zeman & Zydney 2017). Nanofiltration (NF) is a membrane filtration-based method with a pore size of 1–10 nanometers. NF is often used for the treatment of water with low total dissolved solids (TDS) such as surface water and fresh groundwater, for softening and removal of contamination such as natural and synthetic organic matters (Letterman 1999; Roy & Warsinger 2017). RO is also one of the important membrane processes commonly used to remove salt and water salinity as well as total hardness (TH) (Benito & Ruiz 2002; Bodalo-Santoyo *et al.* 2003; Galambos *et al.* 2004). Electrocoagulation is also one of the most effective methods of produced water purification, which has attracted many researchers. This method is mostly used to remove oil and petroleum contaminants in the produced water (Chen *et al.* 2000; Xu & Zhu 2004).

Since each method is capable of separating some specific contaminants, using the purification methods individually is not effective enough. Therefore, combining treatment methods can remarkably increase the quality of water treatment (Baudequin *et al.* 2011; da Silva *et al.* 2015). The quality of the treated water is examined using some factors such as electrical conductivity (EC), chemical oxygen demand (COD), biochemical oxygen demand (BOD_5), total suspended solids (TSS), total dispersed solids (TDS), oil and grease (O&G), total hardness (TH) and turbidity.

The purpose of this study is to design and set up a hybrid pilot-scale wastewater purification system to remove petroleum compounds and water salinity in Iranian offshore oil company wastewater. The treatment system consists of an electrocoagulation section for removing BOD_5 and COD, a microfiltration section to eliminate TSS and following that, a reverse osmosis process to decrease total hardness, salt and salinity. This is a novel hybrid wastewater treatment topology which is effective.

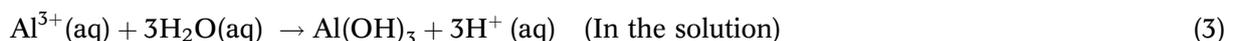
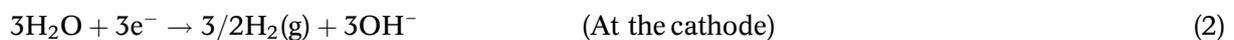
Also, the electrocoagulation reactor is composed of two parts which help to separate flocculation and formed sludge. A comprehensive evaluation of the purified wastewater is carried out by measuring EC, COD, BOD₅, TSS, O&G, TH, DO and pH.

MATERIAL AND METHODS

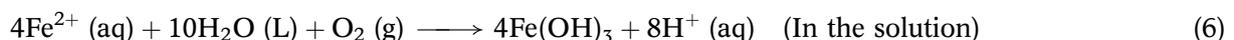
Description of the electrocoagulation process

Electrocoagulation is a treatment method that removes TSS, heavy metals, emulsified oils, bacteria and other contaminants from water. The electrocoagulation is a type of electrolysis process based on multiple reactions due to the transfer of electrons through the soluble interface and electrodes. Several parameters such as the chemistry of the aqueous medium, pH, particle size, and chemical constituent concentrations can influence the electrocoagulation process (Aouni *et al.* 2009). The choice of electrode material in the EC is of particular importance, because each type of electrode is able to remove a number of contaminants with high efficiency. The removal mechanism of heavy metals (Cu, Ni, Zn, and Cr) in the presence of cyanide using Al and Fe electrodes was studied by Kim *et al.* (2020). They reported that using an Fe electrode, higher heavy metal removal is obtained compared to an Al electrode, especially for Cr removal. Rahmani stated that turbidity removal is higher by Al electrodes compared to Fe and St electrodes (Rahmani 2008). Then, removal of BOD and COD using Al and Fe electrodes was investigated by Uğurlu *et al.* (2008). The results showed that the removal of BOD and COD was achieved as 70 and 75% using an Al electrode, while using an Fe electrode the BOD and COD removal was calculated as 80 and 55%, respectively. The reactions of aluminium and iron electrodes are as follows.

For Aluminum electrode



For Iron electrode



The electrocoagulation method is carried out in three steps: First, flocculation is produced by the oxidation of the anode. Then, the pollutants become unstable and, in the third step, the unstable material becomes flocculated and sediments.

Basic principles of ultrafiltration

Ultrafiltration (UF) is a pressure-driven purification process, which is effective in the removal of colloids, proteins, bacteria, pyrogens and other organic molecules larger than 0.1–0.2 μm in size. UF has a lot of applications in many areas such as chemical and pharmaceutical processes, wastewater treatment and the food industry. A large number of UF processes have been commercialized in water and wastewater treatment. Many authors have used UF as wastewater treatment (Li *et al.* 2006; Yan *et al.* 2009) while some have utilized UF as a reverse osmosis pre-treatment (Van Hoof *et al.* 1999;

Petricic *et al.* 2015). Using UF as a pre-treatment for RO can increase water purification quality as well as preventing some drawbacks such as membrane fouling (Wang *et al.* 2011).

Reverse osmosis

Reverse osmosis (RO) is a membrane water purification process that is used to remove ions, organic matter and dissolved and suspended chemical species. RO is an economic process in which there is no need for phase change to remove water. In the RO system, the membrane is the most important and sensitive part of the system, because the required operating pressure is directly related to the thickness and pore size of the membrane. Fouling is the main drawback of RO systems. Therefore, it is important to pre-treat the wastewater before it enters the RO system. RO was used in this study to reduce the wastewater salinity as well as to remove TDS.

EXPERIMENTAL

The produced water characteristics

Produced water was collected from an Iranian offshore oil company, Iran. The produced water was sampled at 2-day intervals and the initial properties were evaluated to compare different analyses in this study. Table 1 shows the characteristics of the three produced water samples.

Table 1 | The characteristics of the three produced water samples

Parameters							
O&G mg/l	Turbidity NTU	TSS mg/l	COD mg/l	BOD ₅ mg/l	EC μs/cm	pH -	Sample
15	176	275	2,300	1,200	4,913	7.47	1
25	70	46	1,960	1,093	4,875	8.34	2
15	191	110	1,815	1,007	4,865	7.15	3

Experimental setup

An experimental pilot-scale system of the designed hybrid wastewater treatment was set up based on Figure 2. The hybrid system is composed of two main sections: electrocoagulation and membrane filtration.

Electrocoagulation section

The electrocoagulation system used in this study is a semi-batch reactor with a volume of 30 liters. The reactor is made of transparent and compact plastic with a size of 40 × 25 × 25 cm. Figure 3 shows the electrocoagulation reactor. The reactor consists of two parts, the lower part containing produced water as well as electrodes, and purified water is collected in the upper part. These two parts are connected by a pipe with a radius of 4.5 cm and water is circulated by the pump in these sections. Two mesh plates are placed at both ends of the connecting pipe. Electrocoagulation reactions occur in the lower part and flocculation is formed during the process, which should be separated effectively. Also, at the end of the electrocoagulation process, sludge is formed on the water surface. The mesh plates help to prevent flocculation and sludge from being transferred to the upper part. So at the end of the process, the time required for the sedimentation of these materials is greatly reduced. In addition,

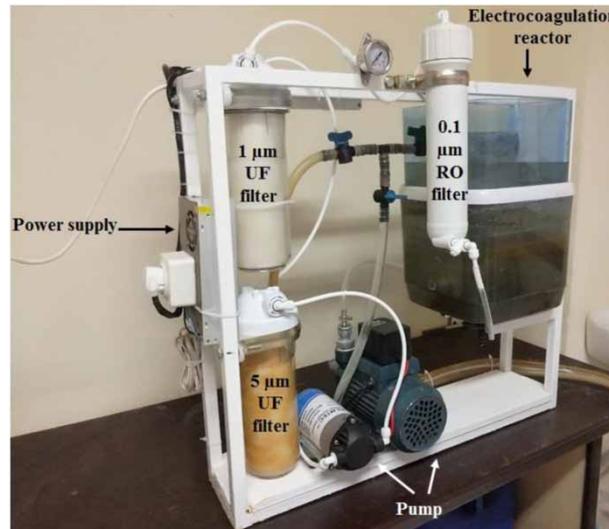


Figure 2 | Experimental setup of the hybrid system.

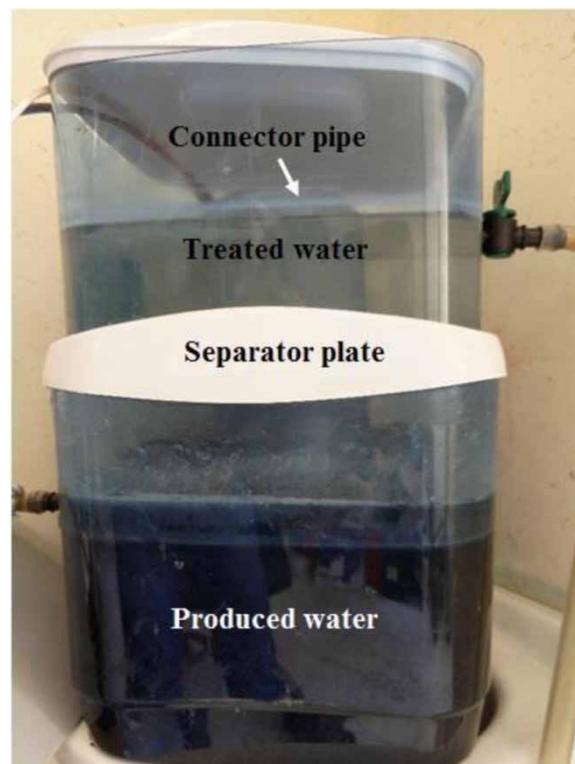


Figure 3 | The electrocoagulation reactor.

during the electrocoagulation process, the pressure in the lower part increases due to the gases released from electrocoagulation reactions. After the electrocoagulation reactions are completed, the amount of produced gases decreases sharply, which leads to a pressure drop in the lower part. In this design, the pump pressure is adjusted so that after the pressure drop of the lower part, the effluent cannot be transferred to the upper part. This helps to determine the completion time of the electrocoagulation reactions. Bhagawan *et al.* suggested that the combination of Al and Fe electrode can lead to higher electrocoagulation efficiency than using them individually (Bhagawan *et al.* 2014). Hence, two iron electrodes as the anode and two aluminium electrodes were used with a size of 12×4 cm and thickness of 2 mm are placed decussate in the lower part of the reactor

2 cm apart. The electrodes were connected to a power supply through four separate wires and the power supply was set to 12 V and current intensity of 15A, 20A and 25A.

Membrane filtration section

In this study, the membrane filtration section is composed of two parts, UF and RO. As previously mentioned, UF is used before the RO process to prevent some drawbacks such as fouling. The UF used in this study consists of two separate membranes in series with a pore size of 5 and 1 μm , respectively. The characteristics of the UF membrane are reported in Table 2. The RO system consists of a membrane with a pore size of 0.1 μm . The characteristics of the RO membrane are reported in Table 3.

Table 2 | Characteristics of the UF membrane

Brand	Model	Process	Pore size	Material
C.C.K.	SC-10-5	UF	5 μm	Polypropylene
C.C.K.	SC-10-1	UF	1 μm	Polypropylene

Table 3 | Characteristics of the RO membrane

Brand	Model	Process	Applied pressure (bar)	Permeate flow rate gpd (l/h)	Stabilized salt rejection (%)
Filmtec	TW30-1812-75	RO	(3.4)	75 (12)	98

Process description

Produced water entered into the electrocoagulation system via a pump. The pollutants were coagulated based on electrocoagulation reactions and precipitated at the bottom of the lower part of the reactor. The treated water was directed to the upper part of the reactor under hydrostatic pressure created by the pump. The purified water was then directed to a 20-litre tank and recycled to the lower part of the reactor to enhance the performance of the electrocoagulation process. This cycle lasted for 300 minutes. After 300 min, the pump was switched off and then treated water was left in the upper part of the reactor for 24 hours to settle the contaminants. It is worth to note that, because the upper part of the reactor is in contact with the environment, the air enters the wastewater and the system cycle. Therefore, no additional airflow is required in this process. Samples were taken at 30 min intervals for analysis. Figure 4 shows a schematic of the electrocoagulation process. The

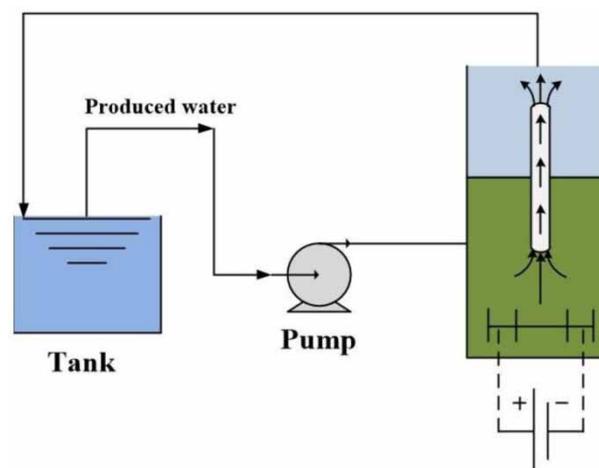


Figure 4 | A schematic of the electrocoagulation process.

electrocoagulation output stream was stored in a 20-litre tank and used for sampling and analysis. The stored water from the electrocoagulation process was directed to the UF part using a pump. After passing the UF process, particles with the size of 1–5 μm were removed and water was directed to the RO system using an RO boosting pump (DOW RO booster pump, 100GPD, 24VDC). The RO system has an input and two outputs. One of the outputs is treated water, which was collected for analyzing and the other one is drain water that was recycled to the storage tank to enhance the purification performance. The water was circulated in the membrane filtration section. [Figure 5](#) depicts the schematic of the filtration section.

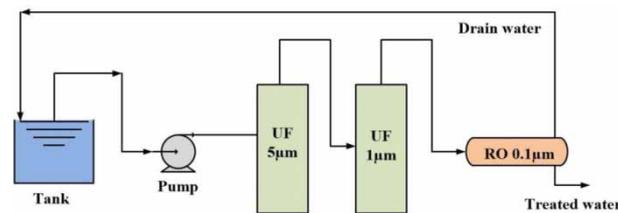


Figure 5 | The schematic of the filtration section.

RESULT AND DISCUSSION

Analytical methods

In order to evaluate the performance of the hybrid wastewater treatment system, various analyses were performed. COD was analyzed using an AQUALYTIC AL800 spectrophotometer. Electrical EC measurements were carried out by JENWAY conductimeter. Turbidity was measured using a TURB WTW353ir turbidimeter. BOD₅ test was performed based on manometric method using BOD₅ Trak (Hach Company). TH was measured using the colorimetric method. YSI 550DO meter was used to measure dissolved oxygen (DO). Oil and grease (O&G) was measured as per the APHA 5520 B standard. TSS was analyzed using the 2540 D standard method.

Hybrid wastewater treatment system

BOD₅ is a factor in determining the amount of oxygen needed for biological stabilization of the organic matter in water. The higher the organic matter in the water, the higher the BOD₅. [Figure 6](#) shows the BOD₅ removal for three produced water samples during the electrocoagulation process. Experimental results showed that the percentage of BOD₅ removal increased significantly over time. In the first 160 min of the electrocoagulation process, the slope of the change in the slope of BOD₅ removal is steep due to the high levels of living microorganisms in the wastewater. The amount of living microorganisms decreases in the wastewater during the time, so the BOD₅ removal tends to almost a constant value. So, it can be said that the highest performance of the electrocoagulation process system occurred in the first 160 min, which led to 82% BOD₅ removal. The final value of BOD₅ removal for samples 1, 2 and 3 reached 91.82%, 91.49% and 90.66%, respectively.

COD is one of the most important indicators of produced water, which represents the amount of organic matter. High COD indicates high organic impurities. [Figure 7](#) illustrates the COD removal of three samples during the electrocoagulation process. Similar to BOD₅ removal, the highest COD removal was observed in the first 160 min, which led to 73% COD removal. The amount of oxidizing materials was high in the early stages of the treatment process. Hence, the slope of the COD removal is expected to be steep at early times. In fact, during the first 160 minutes of the electrocoagulation process, the production of metal ions was increased in the wastewater due to oxidation reactions

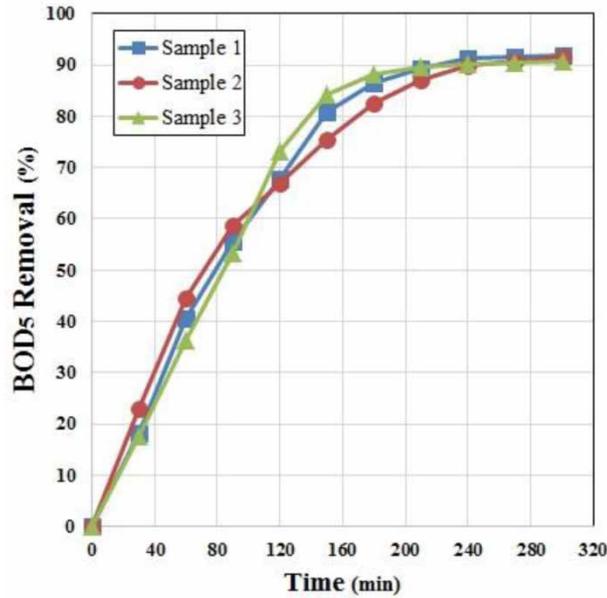


Figure 6 | The BOD₅ removal for three produced water samples during electrocoagulation process.

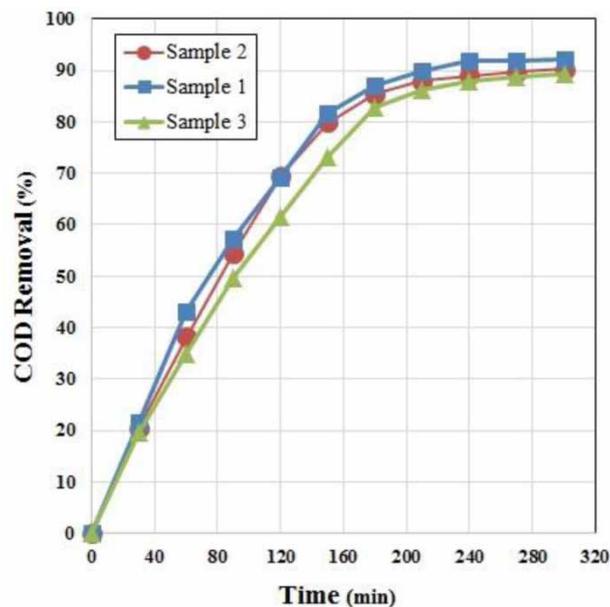


Figure 7 | COD removal percentage of three samples.

and consequently, the coagulation and formation of contaminants' flocculation were increased. Over time, the rates of oxidation reactions decreased due to the reduction of contaminants, which results in a reduction in COD removal. The maximum COD removals for samples 1, 2 and 3 were obtained as 92.17%, 90.25% and 89.36%, respectively.

TSS is a major factor to evaluate the water treatment quality. TSS can be sand, gravel, silt, soil, algae, etc. Figure 8 represents the TSS removal of three samples. During the first 160 min, TSS removed had a steep slope due to the high rate of oxidation reactions and after that, the slope of the TSS removal decreased by decreasing the rate of oxidation due to the removal of pollutants. The results depict that the final TSS removals for samples 1, 2 and 3 were achieved as 88%, 86.95% and 88.18%, respectively.

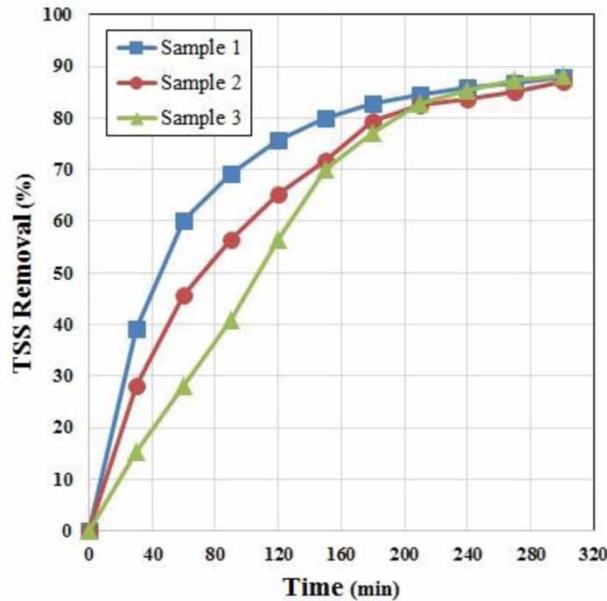


Figure 8 | TSS removal percentage of three samples.

Turbidity is an indicator that represents the clarity of the water as well as water quality. Turbidity is caused by the presence of suspended particles in water. Figure 9 shows seven samples in which samples 1–6 were taken at 50-minute intervals during the electrocoagulation process and sample 7 was collected after the filtration process. As Figure 9 illustrates, at the beginning of the electrocoagulation process the amount of turbidity decreased significantly (samples 1–5) and then increased slightly (sample 6). This is due to the detachment and diffusion of flocculation in the water. Sample 7 depicts that the turbidity has decreased remarkably using filtration systems. Figure 10 illustrates the turbidity removal percentage of three produced water samples. The results show that the maximum turbidity removal percentage was achieved at the 210th min. The maximum turbidity removal percentages for samples 1, 2 and 3 were 66.47%, 28.57% and 71.72%, respectively.



Figure 9 | Samples were taken to determine turbidity.

Oil and grease (O&G) are common pollutants in industrial wastewater, especially in produced water. The removal of O&G is necessary to prevent environmental issues. Figure 11 depicts the O&G removal of three produced water samples during the electrocoagulation process. The results illustrate that the O&G removal increased over time for all samples. At the end of the electrocoagulation process, the O&G removal percentages of samples 1, 2 and 3 were obtained as 65.33%, 80.8% and 66.66%, respectively.

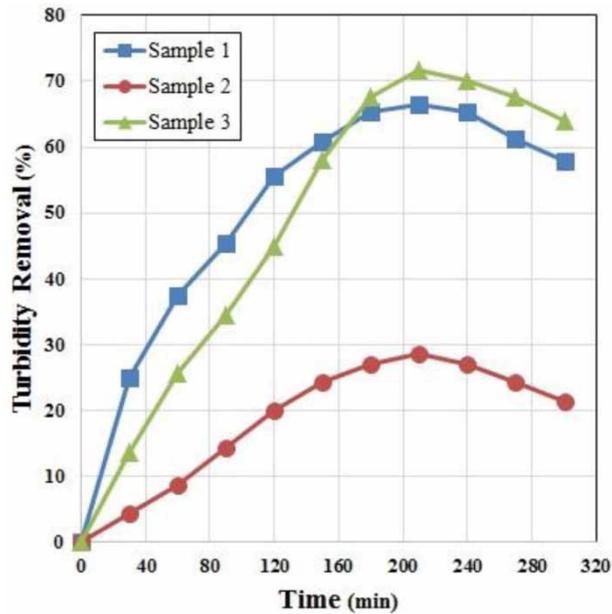


Figure 10 | Turbidity removal percentage of sample 3.

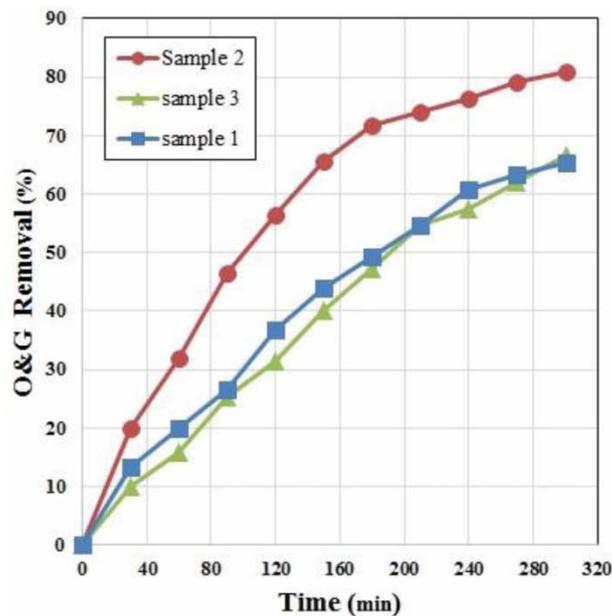


Figure 11 | O&G removal percentage of three samples.

Electrical conductivity (EC) is the ability of water to transmit electric current through free ions. So, EC represents the amount of impurities such as salt and heavy metal ions in water. Most of the free ions are consumed in the electrocoagulation reactions, which leads to a decrease in the impurities' concentration in water. Figure 12 shows the EC for three produced water samples during the electrocoagulation process. At the end of the electrocoagulation process, the EC of samples 1, 2 and 3 decreased from 4,913 $\mu\text{s}/\text{cm}$, 4,875 $\mu\text{s}/\text{cm}$ and 4,865 $\mu\text{s}/\text{cm}$ to 2,654 $\mu\text{s}/\text{cm}$, 2,685 $\mu\text{s}/\text{cm}$ and 2,769 $\mu\text{s}/\text{cm}$, respectively.

The measurement of dissolved oxygen (DO) in wastewater is important because the biological treatment of organic matter requires DO. Figure 13 displays the DO of three produced water samples

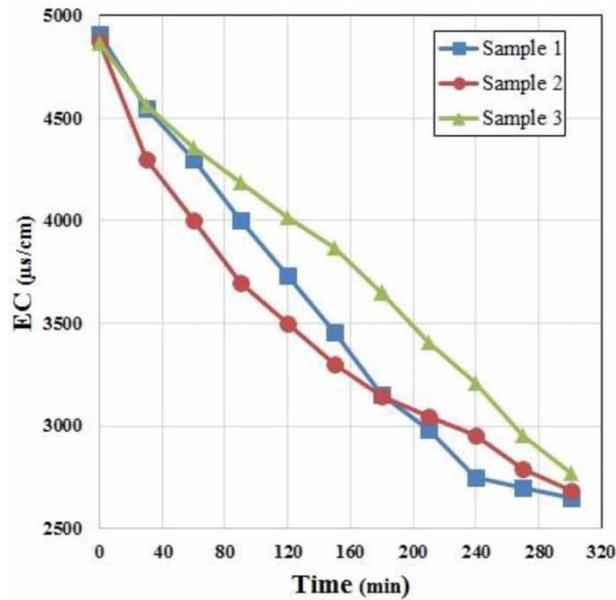


Figure 12 | EC for three produced water samples during the electrocoagulation process.

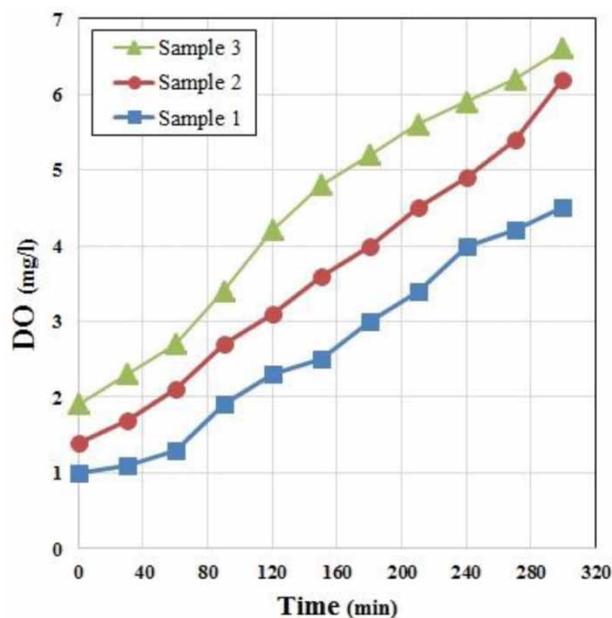


Figure 13 | DO of three produced water samples during electrocoagulation process.

during electrocoagulation process. As [Figure 13](#) shows, the DO of all samples increased over the time for two reasons. Microorganisms help to decompose organic matter by consuming, DO which results in a reduction of wastewater pollution. By the reduction of BOD and COD as well as microorganisms, oxygen consumption reduces significantly, which results in an increase in DO. On the other hand, circulating the wastewater leads to raised DO. Because the upper part of the reactor is in contact with the ambient atmosphere, which causes the DO to rise due to contact with the air. At the end of the electrocoagulation process, the DO of samples 1, 2 and 3 achieved 4.5 mg, 6.2 mg and 6.6 mg, respectively.

Salinity and water total hardness (TH) are two important factors in water treatment. Salinity and TH are caused by dissolved salts in water. To increase water quality, it is necessary to reduce the

TH and salinity of the water to a standard level. Salinity is analyzed based on the EC. The lower the EC, the lower the salinity. Table 4 shows the initial and final values of EC and TH of three produced samples after the membrane filtration process. Results show that the EC of samples 1, 2 and 3 decreased by 89.94%, 89.64% and 89.59%, respectively and the TH of samples 1, 2 and 3 decreased by 83.22%, 86.66% and 82.5%, respectively.

Table 4 | Initial and final values of EC and TH for three produced samples after filtering process

TH		EC		Sample
Final	Initial	Final	Initial	
990	5,900	499	4,913	Sample 1
83.22		89.84		Removal efficiency (%)
1,100	6,000	505	4,875	Sample 2
86.66		89.64		Removal efficiency (%)
1,050	6,000	506	4,865	Sample 3
82.50		89.59		Removal efficiency (%)

Most purification processes, especially chemical and biological processes, are carried out within a specific pH range. The purpose of pH control is not just to neutralize the wastewater: sometimes it is necessary to make the wastewater acidic or basic. The initial and final values of the pH are presented in Table 5 for three produced water samples after the filtration process. Results show that pH for samples 1, 2 and 3 decreased from 6.72, 6.7 and 6.73 to 7.47, 8.34 and 7.15.

Table 5 | PH for three produced water samples after the filtration process

Sample	pH	
	Final	Initial
1	6.72	7.47
2	6.7	8.34
3	6.73	7.15

Effect of electric current

In this study, Response Surface Methodology (RSM) was used to determine the optimal experimental conditions for COD, BOD₅ and turbidity using Design Expert 11. COD, BOD₅ and turbidity were considered to be the variables and the operating parameters were the electrocoagulation process time and electric current. Thirty-three experiments were defined for an examination of the combined operating parameters. Based on the results, the decomposition of aluminium and iron electrodes increased with increasing electric current and, as a result, the concentration of flocculation in water was increased. On the other hand, as the electric current rose, the amount of bubbles produced in the electrocoagulation reactor increased and their diameter decreased. It should be noted that the statistical analysis was done and results are presented in the Appendix. Figure 14 shows the effect of electric current on BOD₅ removal. Results indicate that the optimal electric current and time for BOD₅ removal is 20A and 300 min, respectively.

Figure 15 illustrates the effect of electric current on COD removal. Results depict that the optimal electric current and time for COD removal is 20A and 300 min, respectively.

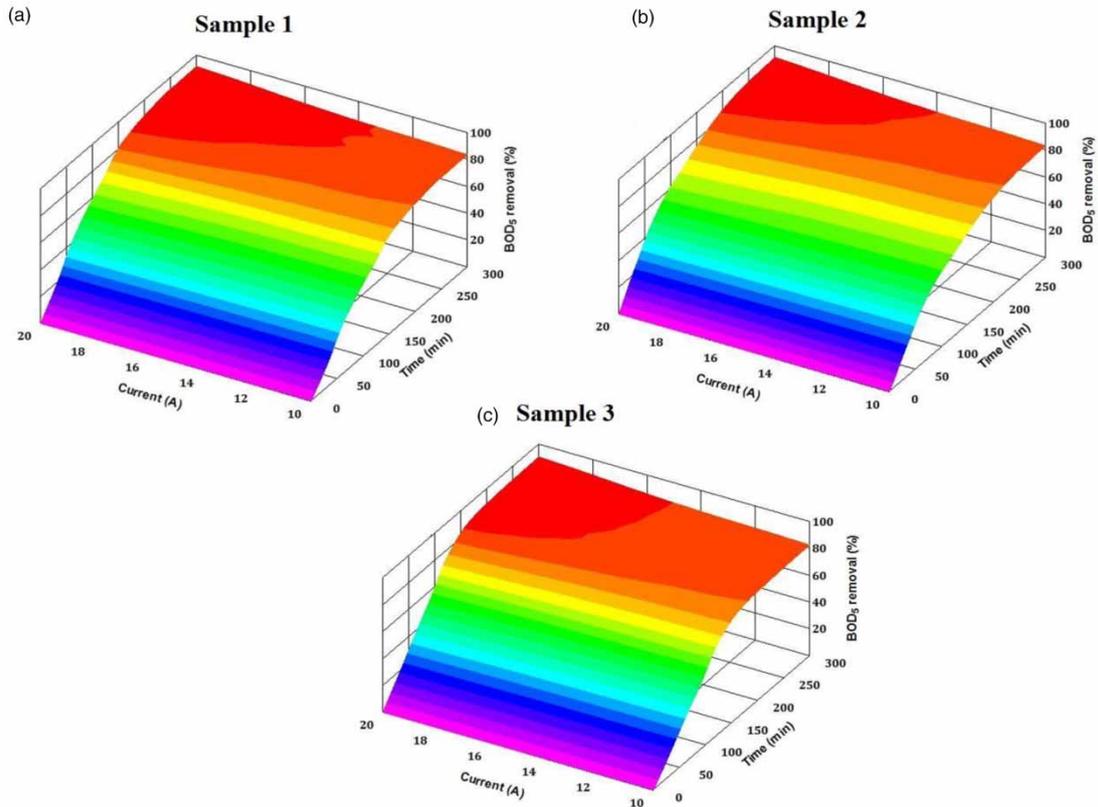


Figure 14 | BOD₅ removal in terms of electric current and time for sample 3.

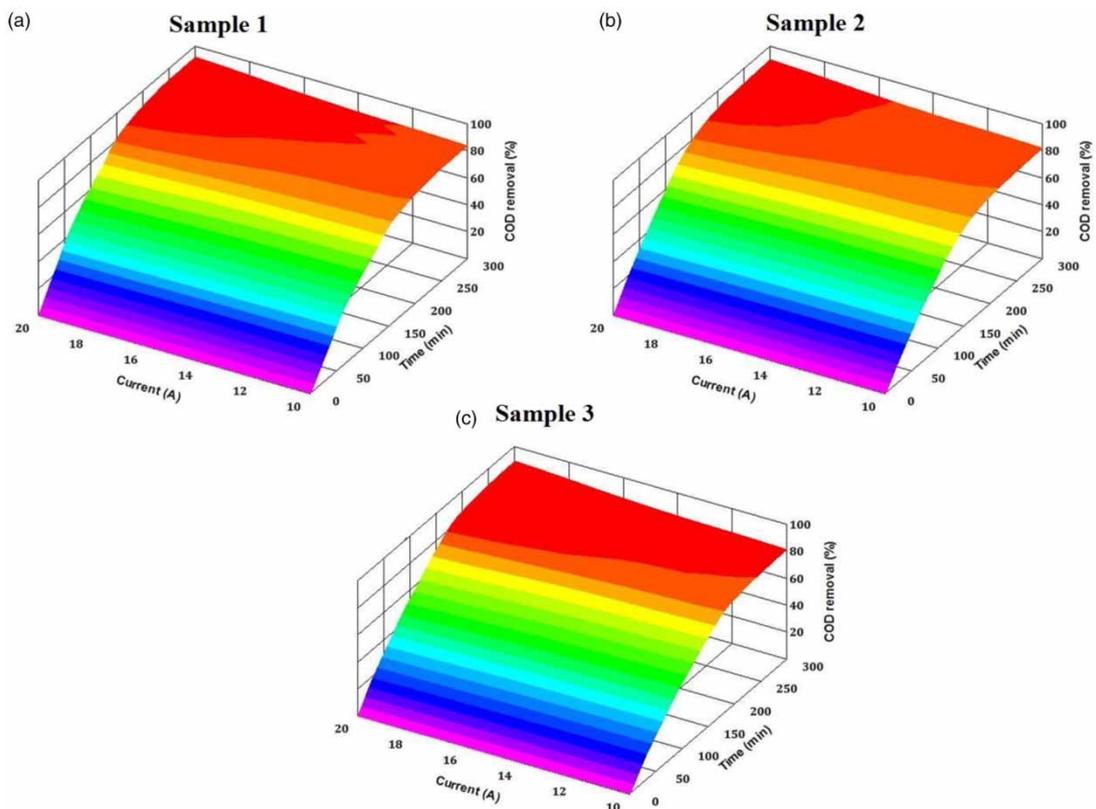


Figure 15 | COD removal in terms of electric current and time for sample 3.

Figure 16 depicts the effect of electric current for turbidity removal. Results illustrate that turbidity removal increased with increasing electric current. As mentioned before, the turbidity decreased with increasing time more than 210 min. So, the optimal electric current and time for turbidity removal are 20A and 210 min, respectively.

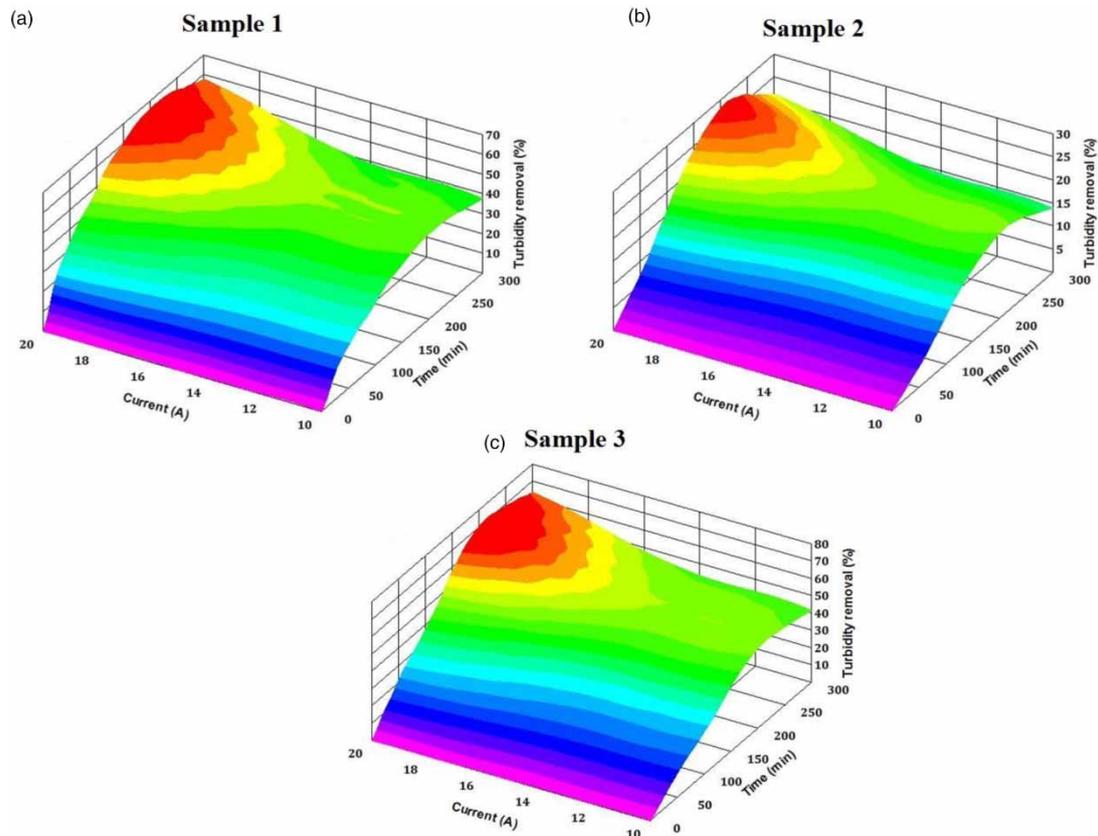


Figure 16 | Turbidity removal in terms of electric current and time for sample 1.

The initial and final values of the studied parameter for treated water are sorted in Table 6. Based on Table 6, the hybrid treatment system in this study has acceptable performance in produced water treatment.

Table 6 | The initial and final values of the studied parameters for treated water

Sample	Sample 1		Sample 2		Sample 3	
	Initial	Final	Initial	Final	Initial	Final
BOD ₅ (mg/l)	1,200	98.1	1,093	93	1,007	94
COD (mg/l)	2,300	180	1,960	191	1,815	193
TSS (mg/l)	275	33	46	6	110	13
EC (μs/cm)	4,913	499	4,875	505	4,865	506
O&G (mg/l)	15	5.2	25	4.8	15	5
PH	7.47	6.72	8.34	6.7	7.15	6.73
Turbidity (NTU)	176	74	70	55	191	69
TH (mg/l)	5,900	990	6,000	1,100	6,000	1,050
DO (mg/l)	1	4.5	1.4	6.2	1.9	6.6

CONCLUSION

In this study, a hybrid treatment system consisting of electrocoagulation, ultrafiltration (UF) and reverse osmosis (RO) sections was used to purify produced water. Three samples of Iranian offshore oil company produced water were used to examine the performance of the hybrid system. In the electrocoagulation section, iron and aluminum were applied as the electrodes with an electric current of 15A, 20A and 25A. Two membranes with a pore size of 1 and 5 μm in the UF section and a membrane with a pore size of 0.1 μm in the RO section were employed. Results illustrated that the maximum final removal efficiencies of BOD₅, COD, TSS, O&G, EC, TH and turbidity were obtained as 94.49%, 92.17%, 88.18%, 80.8%, 89.84%, 86.66% and 71.72%, respectively. Also, the maximum DO was achieved as 6.6 mg. Response Surface Methodology (RSM) was used to determine the optimal experimental conditions for COD, BOD₅ and turbidity using Design Expert11. RSM results depicted that the optimal electric current and time for BOD₅, COD and turbidity removal is 20A and 300 min, respectively. The study outcomes proved that the designed hybrid system has a great performance and treated water has acceptable quality.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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